研 究

Mutual Relation between Distributions Based on Number and Mass
in The Particle Size-Shape Dispersion Diagram

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SYNOPSIS

In order to investigate the correlation between number and mass bases of both the particle size and shape, the particle size-shape dispersion diagrams were used. On the assumption that the dispersion condition between particle size and shape is expressed with the two dimensional normal distribution laws, the theoretical consideration of the conversion between number and mass bases revealed the equations expressing the correlation between the two bases. The equations of basis conversion derived in this study contain Hatch's equations for the particle size distribution. The average value of the shape distribution changed with the standard deviations of both size and shape distributions and the correlation coefficient between them.

KEY WORDS

particle size, particle shape, particle size-shape dispersion diagram, basis conversion

List of symbols

- C_s symbol defined as $C_s \equiv \sigma_s \sqrt{1 - r_{sz}^2}$
 C_z symbol defined as $C_z \equiv \sigma_z \sqrt{1 - r_{sz}^2}$
 D_p particle diameter (projected area diameter)
 D_{50} diameter corresponding to 50% of cumulative frequency
 p probability frequency or probability density
 r_{sz} correlation coefficient between two parameters, S and Z
 S shape parameter defined as $\ln\{\Psi_c/(1-\Psi_c)\}$
 V_p particle volume
 Z size parameter defined as $\ln(D_p)$
 ϕ_v volume shape factor
 ρ particle density
 σ_s standard deviation about shape parameter S
 σ_z standard deviation about size parameter Z
 Ψ_c circularity of a projected particle
 Ψ_{50} circularity corresponding to 50% of cumulative frequency
- Superscripts**
 $-$ average value (ex. $\bar{S}$ and $\bar{Z}$)
 o number basis (ex. C_s^o , C_z^o , D_{50}^o , p^o , r_{sz}^o , $\bar{S}^o$, $\bar{Z}^o$, σ_s^o , σ_z^o and Ψ_{50}^o)
 $^{\#}$ mass basis (ex. $C_s^{\#}$, $C_z^{\#}$, $D_{50}^{\#}$, $p^{\#}$, $r_{sz}^{\#}$, $\bar{S}^{\#}$, $\bar{Z}^{\#}$, $\sigma_s^{\#}$, $\sigma_z^{\#}$ and $\Psi_{50}^{\#}$)

1 Introduction

Particle size distribution has been determined as a powder property in vast powder industry. There can be various particle size distributions based on different measuring methods selected¹⁾. Most popular ones are the number- and mass-based distributions in particle size. Then mutual relation between the two distributions is necessary for conversion needed in comparison among the ones measured on the different bases, and/or in consideration for the

designing of blending the powders that have the size distributions given on the number basis.

Two distribution laws of log-normal and Rosin-Rammler have been widely used for expressing the particle size distribution. In the log-normal distribution law, the relationship between the number and mass bases has been expressed by Hatch's equations derived analytically²⁾. The similar relation in the Rosin-Rammler distribution law has been reported in the previous work³⁾. The relations in these

laws can be valid under the assumption that all particles in a powder are of a same shape. In actual powder, however, there is a particle shape distribution. Then the above assumption has no reality.

In the present work, the mutual correspondence between the number- and mass-based distributions is discussed by using some correlation between the size and shape distributions in the particle size-shape dispersion diagram proposed for more precise characterization⁴⁻⁹. It has been proposed that the dispersion condition between particle size and shape in the diagram can be expressed with a two-dimensional normal distribution law. The theoretical conversion between the number and mass bases in the above diagram is studied and the conversing equations are derived. The results of the basis conversion are also shown by changes in the dispersion diagram.

2 Particle size-shape dispersion diagram and two-dimensional normal distribution law

The previous reports have already shown the characteristics of the particle size-shape dispersion diagram⁴, and some quantitative relations between the characterized powder properties and the behavior, e.g. the packing density⁵⁻⁸ or the fluidity⁹. Here the diagram and the two-dimensional normal distribution law are briefly summarized.

An example of the particle size-shape dispersion diagram is shown in Fig.1. The axes of abscissas and ordinates are expressed with the particle diameter D_p (projected area diameter) and the circularity Ψ_c that is one of the shape indexes, respectively. This diagram is composed of the size frequency distribution (A), the shape frequency distribution (B) and the two-dimensional figure (C), indicating the dispersion condition between the size and shape. The two-dimensional figure in the diagram made on the number basis is usually characterized by plotting the cross points which correspond to the coordinates of size and shape in the powder particles. It can also be expressed with the iso-probability density curves as shown in Fig.1. The particle size-shape dispersion diagram can be used for numerical characterization of powder characteristics according to the distributions of particle size and shape and the correlation between them.

The axis of particle size in the diagram is on a logarithmic scale, that is $\ln(D_p)$, and the other axis of particle shape is scaled with $\ln\{\Psi_c/(1-\Psi_c)\}$. These styles of axes are based on the condition that the size and shape distributions obey the normal distribution laws having the parameters $\ln(D_p)$ and $\ln\{\Psi_c/(1-\Psi_c)\}$, respectively⁴. Thus the dispersion

figure in this diagram is an ideal condition that obeys the two-dimensional normal distribution law on the number basis. In this case the number-based size distribution (A) is expressed by the following equation,

$$p^o(Z) = \frac{1}{\sqrt{2\pi}\sigma_z^o} \exp\left\{-\frac{(Z-\bar{Z}^o)^2}{2\sigma_z^{o2}}\right\}, \quad (1)$$

where $p^o(Z)$ is the probability frequency, Z the size parameter defined as $Z \equiv \ln(D_p)$, σ_z^o the standard deviation about parameter Z , and $\bar{Z}^o$ the average value of parameter Z or the logarithmic value of the diameter corresponding to 50% of cumulative frequency, D_{50}^o . The particle shape distribution of number basis (B) is similarly expressed as follows:

$$p^o(S) = \frac{1}{\sqrt{2\pi}\sigma_s^o} \exp\left\{-\frac{(S-\bar{S}^o)^2}{2\sigma_s^{o2}}\right\}. \quad (2)$$

The symbols $p^o(S)$, σ_s^o and $\bar{S}^o$ are the probability frequency, the standard deviation, and the average value about the shape parameter S defined as $S \equiv \ln\{\Psi_c/(1-\Psi_c)\}$, respectively. $\bar{S}^o$ can be defined as $\bar{S}^o \equiv \ln\{\Psi_{50}^o/(1-\Psi_{50}^o)\}$, where Ψ_{50}^o is the circularity corresponding to 50% of cumulative frequency. Furthermore, the number-based dispersion figure (C) between the size and shape distributions is expressed with the two-dimensional normal distribution law,

Fig.1 Particle size-shape dispersion diagram based on the two-dimensional normal distribution law; symbols A, B and C indicate the size frequency distribution, the shape frequency one and the two-dimensional figure indicating the dispersion condition between size and shape, respectively.

($D_{50}^o=5\mu\text{m}$, $\sigma_z^o=0.5$, $\Psi_{50}^o=0.8$, $\sigma_s^o=0.5$, $r_{sz}^o=-0.4$)

$$p^o(S, Z) = \frac{\sqrt{1-r_{sz}^2}}{2\pi C_s^o C_z^o} \exp \left[-\frac{1}{2} \left\{ \frac{(S-\bar{S}^o)^2}{C_s^{o2}} - 2r_{sz}^o \frac{(S-\bar{S}^o)(Z-\bar{Z}^o)}{C_s^o C_z^o} + \frac{(Z-\bar{Z}^o)^2}{C_z^{o2}} \right\} \right] \quad (3)$$

where $p^o(S, Z)$ and r_{sz}^o are the probability density and the correlation coefficient between the two parameters, S and Z . The symbols C_s^o and C_z^o are defined as $C_s^o \equiv \sigma_s^o \sqrt{1-r_{sz}^{o2}}$ and $C_z^o \equiv \sigma_z^o \sqrt{1-r_{sz}^{o2}}$.

There are three types of diagram according to the correlation coefficient r_{sz}^o . The examples are indicated in Fig.2 showing iso-probability curves with correlation coefficients of negative, zero, and positive values, respectively. Under the condition that the correlation coefficient has a negative value ($r_{sz}^o = -0.4$) as shown in Fig.2(a), the powder has a tendency for the irregularity of the particle shape to increase with the particle size. Many powders are classified into this type. Fig.2(b) indicates the condition when $r_{sz}^o = 0$. There is no correlation between size and shape under this condition, i.e. both distributions are independent of each other. The condition indicated in Fig.2(c) is another one, that is, the correlation coefficient has a positive value ($r_{sz}^o = 0.4$). In this case the powder has an opposite tendency to that shown in Fig.2(a).

3 Derivations of Hatch's equations on the size distribution and basis conversion equation on the size-shape dispersion diagram

Hatch's equations are well known as the conversion equations between number and mass bases of the particle size distribution that obeys the log-normal distribution law. Its theoretical derivation is summarized as follows:

The mass-based probability frequency p^* of the particle size distribution is generally connected with the frequency p^o on the number basis according to the following equation,

$$p^* \equiv \frac{\rho \cdot \phi_v \cdot D_p^3 \cdot p^o}{\int_{D_p=0}^{\infty} (\rho \cdot \phi_v \cdot D_p^3 \cdot p^o) dZ} \quad (4)$$

This equation is obtained by normalizing the product between the number-based probability frequency p^o and the mass of a particle $\rho \cdot \phi_v \cdot D_p^3$ with its integration. The symbols ρ and ϕ_v are the particle density and the volume shape factor, respectively. Assuming particles of a same density and a similar shape, Eq. (4) can be simplified into the equation,

$$p^* \equiv \frac{D_p^3 \cdot p^o}{\int_{D_p=0}^{\infty} (D_p^3 \cdot p^o) dZ} \quad (5)$$

Fig.2 Typical particle size-shape dispersion diagrams under three different conditions: (a) $r_{sz}^o = -0.4$; (b) 0; and (c) 0.4.
($D_{50} = 5\mu\text{m}$, $\sigma_z^o = 0.5$, $\Psi_{50}^o = 0.8$, $\sigma_s^o = 0.5$)

For the probability frequency p^o of the number-based size distribution expressed with Eq. (1), the above equation becomes

$$p^*(Z) = \frac{\frac{1}{\sqrt{2\pi}\sigma_z^o} \exp\left\{-\frac{(Z-\bar{Z}^o)^2}{2\sigma_z^{o2}} + 3Z\right\}}{\int_{-\infty}^{\infty} \frac{1}{\sqrt{2\pi}\sigma_z^o} \exp\left\{-\frac{(Z-\bar{Z}^o)^2}{2\sigma_z^{o2}} + 3Z\right\} dZ} \quad (6)$$

After arrangement of the above equation,

$$p^*(Z) = \frac{\frac{D_{50}^{o3}}{\sqrt{2\pi}\sigma_z^o} \exp\left(\frac{9}{2}\sigma_z^{o2}\right) \exp\left\{-\frac{(Z-\bar{Z}^o-3\sigma_z^{o2})^2}{2\sigma_z^{o2}}\right\}}{\int_{-\infty}^{\infty} \frac{D_{50}^{o3}}{\sqrt{2\pi}\sigma_z^o} \exp\left(\frac{9}{2}\sigma_z^{o2}\right) \exp\left\{-\frac{(Z-\bar{Z}^o-3\sigma_z^{o2})^2}{2\sigma_z^{o2}}\right\} dZ} \quad (7)$$

is obtained. This equation is simplified as follows:

$$p^*(Z) = \frac{\frac{1}{\sqrt{2\pi}\sigma_z^o} \exp\left\{-\frac{(Z-\bar{Z}^o-3\sigma_z^{o2})^2}{2\sigma_z^{o2}}\right\}}{\int_{-\infty}^{\infty} \frac{1}{\sqrt{2\pi}\sigma_z^o} \exp\left\{-\frac{(Z-\bar{Z}^o-3\sigma_z^{o2})^2}{2\sigma_z^{o2}}\right\} dZ} \quad (8)$$

Since the denominator in this equation is infinite integral of the normal distribution that has the average value $\bar{Z}^o + 3\sigma_z^{o2}$ and the standard deviation σ_z^o , its value becomes unity. The probability frequency p^* of the mass-based size distribution becomes

$$p^*(Z) = \frac{1}{\sqrt{2\pi}\sigma_z^o} \exp\left\{-\frac{(Z-\bar{Z}^o-3\sigma_z^{o2})^2}{2\sigma_z^{o2}}\right\} \quad (9)$$

By comparing Eqs. (1) and (9), it is clear that the converted distribution equation of mass basis has the same style with the number-based distribution obeying normal distribution law. Then we have the following basis conversion equations:

$$\bar{Z}^* = \bar{Z}^o + 3\sigma_z^{o2} \text{ and } \sigma_z^* = \sigma_z^o \quad (10a)$$

or

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$$D_{50}^* = D_{50}^o \cdot \exp(3\sigma_z^{o2}) \text{ and } \sigma_z^* = \sigma_z^o \quad (10b)$$

These are called Hatch's equations.

An attempt is made in the similar manner to derive the basis conversion equation of the particle size-shape dispersion diagram. The probability density p^* of the mass-based dispersion diagram can be expressed with following equation in place of Eq. (4)

$$p^* = \frac{\rho \cdot \phi_v \cdot D_p^3 \cdot p^o}{\int_{D_p=0}^{\infty} \int_{\psi_c=0}^1 (\rho \cdot \phi_v \cdot D_p^3 \cdot p^o) dS \cdot dZ} \quad (11)$$

where the symbol p^o means the probability density of the number-based dispersion diagram. Assuming particle of a same material, the particle density ρ is independent of both the particle size and shape. The volume shape factor ϕ_v is defined as V_p/D_p^3 , where the symbol V_p is the particle volume. Generally, the shape factor ϕ_v is dependent on the particle shape. When the equivalent volume diameter defined as $\sqrt[3]{6V_p/\pi}$ is used as the particle size D_p , the shape factor ϕ_v becomes the constant value of $\pi/6$ inevitably. The projected area diameter is used as D_p in the number-based dispersion diagram. Assuming that the projected area diameter is proportional to the equivalent volume diameter, the volume shape factor ϕ_v becomes a constant value independent of both the particle size and shape. Thus, the following equation can be derived.

$$p^* = \frac{D_p^3 \cdot p^o}{\int_{D_p=0}^{\infty} \int_{\psi_c=0}^1 (D_p^3 \cdot p^o) dS \cdot dZ} \quad (12)$$

For the probability density p^o of the number-based dispersion condition expressed with Eq. (3), the numerator in the above equation becomes

$$D_p^3 \cdot p^o = \frac{\sqrt{1-r_{sz}^{o2}}}{2\pi C_s^o C_z^o} \exp \left[-\frac{1}{2} \left\{ \frac{(S-\bar{S}^o)^2}{C_s^{o2}} - 2r_{sz}^o \frac{(S-\bar{S}^o)(Z-\bar{Z}^o)}{C_s^o C_z^o} + \frac{(Z-\bar{Z}^o)^2}{C_z^{o2}} \right\} + 3Z \right] \quad (13)$$

After arrangement of the above equation, the following equation is obtained.

$$D_p^3 \cdot p^o = D_{50}^{o3} \exp\left(\frac{9}{2}\sigma_z^{o2}\right) \frac{\sqrt{1-r_{sz}^{o2}}}{2\pi C_s^o C_z^o} \exp \left[-\frac{1}{2} \left\{ \frac{(S-\bar{S}^o-3r_{sz}^o\sigma_s^o\sigma_z^o)^2}{C_s^{o2}} - 2r_{sz}^o \frac{(S-\bar{S}^o-3r_{sz}^o\sigma_s^o\sigma_z^o)(Z-\bar{Z}^o-3\sigma_z^{o2})}{C_s^o C_z^o} + \frac{(Z-\bar{Z}^o-3\sigma_z^{o2})^2}{C_z^{o2}} \right\} \right] \quad (14)$$

The above equation contains the two-dimensional normal distribution that has the two average values $\bar{S}^o + 3r_{sz}^o\sigma_s^o\sigma_z^o$ and $\bar{Z}^o + 3\sigma_z^o$, the two standard deviations σ_s^o and σ_z^o , and the correlation coefficient r_{sz}^o . The denominator in Eq. (12) corresponds to the double integral of Ep. (14). Since the infinite integral of the two-dimensional normal

distribution becomes unity, the denominator in Eq. (12) can be expressed with the following equation.

$$\int_{D_p=0}^{\infty} \int_{\Psi_c=0}^1 (D_p^3 \cdot p^o) dS \cdot dZ = D_{s0}^3 \exp\left(\frac{9}{2}\sigma_z^{o2}\right) \quad (15)$$

The probability density p^* of the mass-based dispersion diagram can be derived.

$$p^*(S, Z) = \frac{\sqrt{1-r_{sz}^{o2}}}{2\pi C_s^o C_z^o} \exp\left[-\frac{1}{2}\left\{\frac{(S-\bar{S}^o-3r_{sz}^o\sigma_s^o\sigma_z^o)^2}{C_s^{o2}} - 2r_{sz}^o \frac{(S-\bar{S}^o-3r_{sz}^o\sigma_s^o\sigma_z^o)(Z-\bar{Z}^o-3\sigma_z^o)}{C_s^o C_z^o} + \frac{(Z-\bar{Z}^o-3\sigma_z^o)^2}{C_z^{o2}}\right\}\right] \quad (16)$$

Fig.3 Relationship between number and mass bases on particle size and shape distributions under three different conditions of correlation coefficient: (a) shows particle size distribution and (b)–(d) show particle shape distributions of: (b) $r_{sz}^o = -0.4$; (c) 0; and (d) 0.4, respectively. ($D_{s0}^o = 5\mu\text{m}$, $\sigma_z^o = 0.5$, $\Psi_{s0}^o = 0.8$, $\sigma_s^o = 0.5$)

By comparing Eqs. (3) and (16), it is clear that the converted distribution equation of mass basis has the same style with the number-based one obeying two-dimensional normal distribution law. Then we have the following basis conversion equations;

$$\begin{aligned}\bar{S}^* &= \bar{S}^o + 3r_{sz}^o \sigma_s^o \sigma_z^o, & \sigma_s^* &= \sigma_s^o \\ \bar{Z}^* &= \bar{Z}^o + 3\sigma_z^{o^2}, & \sigma_z^* &= \sigma_z^o \\ \text{and} & & r_{sz}^* &= r_{sz}^o\end{aligned}\quad (17a)$$

or

$$\begin{aligned}\frac{\Psi_{s0}^*}{1 - \Psi_{s0}^*} &= \frac{\Psi_{s0}^o}{1 - \Psi_{s0}^o} \exp(3r_{sz}^o \sigma_s^o \sigma_z^o), & \sigma_s^* &= \sigma_s^o \\ D_{s0}^* &= D_{s0}^o \cdot \exp(3\sigma_z^{o^2}), & \sigma_z^* &= \sigma_z^o \\ \text{and} & & r_{sz}^* &= r_{sz}^o\end{aligned}\quad (17b)$$

Various interesting information can be obtained from these equations. The basis conversion equations relating to the particle size distribution are in agreement with Hatch's equations. The correlation between the particle shape distributions (especially the average values of the distribution) of both bases, varies according to the standard deviations of both the size and shape distributions and the correlation coefficient indicating the dispersion condition. The standard deviation of the shape distribution and the correlation coefficient, are the same.

The cumulative and relative frequency distributions of particle size and shape on both bases are shown in Fig.3. The cumulative distributions are plotted on the normal probability diagrams. The relative frequency distribution is defined as the frequency distribution standardized by the maximum frequency. The solid and broken lines in these figures indicate the distributions based on number and mass, respectively. As for the particle size distribution shown in Fig.3(a), two straight lines, which are parallel in the cumulative frequency distribution diagram, indicates that the standard deviations of both the distributions have the same value. On the other hand, Fig.3(b),(c) and (d) show three types of particle shape distributions according to the correlation coefficient r_{sz}^o . In the case of $r_{sz}^o < 0$ as shown in Fig.3(b), the broken straight line of mass basis slides from the solid straight line of number basis, toward the left side, that is, to a smaller average value of the shape distribution. In the case of no correlation $r_{sz}^o = 0$ as shown in Fig.3(c), the mass-based shape distribution is in agreement with the number-based one. Fig.3(d) shows the opposite condition ($r_{sz}^o > 0$) to Fig.3(b). The broken straight line of the mass basis moves to a larger average value of the shape distribution. The standard deviations keep the same value in all cases.

The three types are also shown in Fig.4(a)-(c) of the particle size-shape dispersion diagrams. These figures indicate the results obtained under the same number-based conditions, except to the correlation coefficient r_{sz}^o , i.e. Fig.4(a) is the case of negative correlation ($r_{sz}^o = -0.4$), (b) is the case of non-correlation ($r_{sz}^o = 0$), and (c) is the case of positive correlation. The solid and broken lines are respectively based on the number and mass bases the same as in Fig.3. The mutual relations between bases of the size distribution are the same in these three dispersion diagrams. It means that they don't depend on the correlation coefficient r_{sz}^o . In the conversion of number basis to mass basis, the dispersion condition between the size and shape in the diagram moves toward the right and down, the right, or the right and up, which depends upon the negative correlation, non-correlation, or positive correlation, respectively. This means that the dispersion condition is affected by the basis conversion.

The adequacy of Hatch's equations (10) can be easily confirmed by comparison between results of number- and mass-based size distribution analyses, e.g. microscopic method and sieve method. Since mass-based size-shape dispersion analysis has not been established yet, the adequacy of the basis conversion equations (17), which can be derived theoretically, can be hardly confirmed by measurement. The confirmation is possible as the classifying-shape technology develops. The theoretical process deriving the basis conversion equations (17), however, can not be used only for the size-shape dispersion diagram of powder, but also for the diagrams of all discrete materials, e.g. the short fiber which has been reported in the previous works^{10,11}.

4 Conclusion

In this work the particle size-shape dispersion diagrams are used in order to investigate the correlation between number and mass bases according to both the particle size and shape. On the assumption that the dispersion condition between particle size and shape is expressed with the two-dimensional normal distribution laws, the theoretical consideration of the conversion between number and mass bases reveals the equations expressing the correlation between both the bases. The variations on the dispersion diagram are also researched according to the conversion equations.

The results are summarized as follows:

1. The basis conversion equations with respect to the particle size distribution are consistent with Hatch's equations. The standard deviation of the distribution does not change with the conversion of number basis

Fig.4 Three types of particle size-shape dispersion diagram based on both of number (solid line) and mass (broken line) bases: (a) $r_{s2} = -0.4$; (b) 0; and (c) 0.4.
($D_{50} = 5\mu\text{m}$, $\sigma_z^2 = 0.5$, $\Psi_{50}^0 = 0.8$, $\sigma_s^2 = 0.5$)

into mass basis. On the other hand, the average value of the distribution increases with the conversion.

2. The standard deviation of the shape distribution does not changed with the conversion of number basis into mass basis the same as that of the size distribution. The average value of the distribution, however, changes with the standard deviations of both size and shape distributions and the correlation coefficient between them.
3. In the conversion of number basis to mass basis, the dispersion condition between the size and shape in the diagram moves toward the right down, the right and the right up according to the negative correlation, non-correlation and positive correlation, respectively.
4. The theoretical process deriving the basis conversion equations can not be used only for the size-shape dispersion diagram of powder, but also for the diagrams of all discrete materials.

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